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Ground-based FTIR measurements of CO from the Jungfraujoch: characterisation and comparison with in situ surface and MOPITT data

B. Barret¹, M. De Mazière¹, and E. Mahieu²

¹Belgian Institute for Space Aeronomy, Brussels, Belgium

²Institut de Géophysique et d'Astrophysique, Liège, Belgium

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Abstract. CO vertical profiles have been retrieved from solar absorption FTIR spectra recorded at the NDSC station of the Jungfraujoch (46.5° N, 8° E and 3580 m a.s.l.) for the period from January 1997 to May 2001. The characterisation of these profiles has been established by an information content analysis and an estimation of the error budgets. A partial validation of the profiles has been performed through comparisons with correlative measurements. The average volume mixing ratios (vmr) in the 3 km layer above the station have been compared with coincident surface measurements. The agreement between monthly means from both measurement techniques is very good, with a correlation coefficient of 0.87, and no significant bias observed. The FTIR total columns have also been compared to CO partial columns above 3580 m a.s.l. derived from the MOPITT (Measurement Of Pollution In The Troposphere) instrument for the period March 2000 to May 2001. Relative to the FTIR columns, the MOPITT partial columns exhibit a positive bias of $8 \pm 8\%$ for daytime and of $4 \pm 7\%$ for nighttime measurements.

1 Introduction

As it is responsible for about 75% of the OH radical sink, carbon monoxide (CO) plays a major role in atmospheric chemistry (Thompson, 1992). It affects the radiative forcing of the atmosphere by its influence on the concentrations of greenhouse gases such as CH₄ and O₃ (Daniel and Solomon, 1998). Space-borne instruments providing quasi-continuous and global observations of CO are regarded as highly valuable for inverse modeling to infer its natural and anthropogenic sources (Bergamaschi et al., 2000). Their validation by independent observations is therefore of great importance. Ground-based FTIR (Fourier Transform Infrared) instru-

ments provide accurate measurements of total columns of many atmospheric trace gases. Profile retrieval algorithms improve the quality of the total column measurements while providing information on the vertical distributions. We present a characterisation and a partial validation of the retrieved ground-based FTIR profiles of CO at the Network for the Detection of Stratospheric Changes (NDSC) International Scientific Station of the Jungfraujoch (ISSJ) in the Swiss Alps. The characterisation includes an information content analysis and a determination of the error budgets. The validation is performed through comparisons between the FTIR retrieved “surface” (3.58–6.5 km) vmr and correlative surface in situ measurements. We also present comparisons between CO columns and profiles from the FTIR retrievals with MOPITT partial columns.

2 Characterisation of CO profiles retrieved from ground-based FTIR at ISSJ

2.1 Retrieval parameters

CO height resolved information has been retrieved from ground-based solar absorption FTIR spectra by e.g. Rinsland et al. (2000) at the ISSJ and Jones et al. (2001) at the NDSC station at Lauder, New Zealand. In the present study, we use the same CO absorption lines and algorithm (SFIT2) as in these two studies. The spectroscopic parameters for the CO absorption lines are taken from the HITRAN2000 database (Rothman et al., 2003). The CO a priori profile used for the retrieval is the same as in the MOPITT version 3 retrievals (Deeter et al., 2003). The CO concentrations are retrieved in 29 layers from the ground (3.58 km) up to 100 km. The first retrieval layer extends from 3.58 to 4.5 km and, from 4.5 to 20.5 km, the retrieval layers are 1 km thick.

Correspondence to: B. Barret
(bbarret@ulb.ac.be)

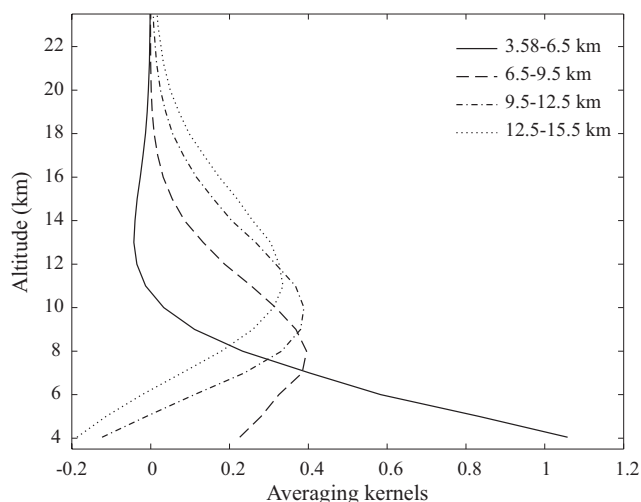


Fig. 1. CO vmr averaging kernels for 3 km thick merged layers from the ground up to 15.5 km. They have been calculated for a SZA (Solar Zenith Angle) of 61° and an OPD (Optical Path Difference) of 175 cm.

In the algorithm SFIT2 (Rinsland et al., 1998), the retrievals are based on a semi-empirical implementation of the Optimal Estimation Method (OEM) (Rodgers, 1976). The a priori and the measurement covariance matrices are ad hoc matrices chosen so that the best possible fit is obtained without undesired oscillations in the retrieved profiles. The ad hoc CO a priori covariance matrix (S_x) has diagonal values corresponding to standard deviations of 40% and 30% in the two lowermost layers and of 25% above. In order to account for correlations between CO values at different altitudes (z , in km), extra-diagonal terms are added to S_x as gaussian functions: $S_{x_{ij}} = \sqrt{S_{x_{ii}} S_{x_{jj}}} \exp(-\frac{z_i - z_j}{2})^2$. The ad hoc measurement covariance matrix is diagonal with values corresponding to a signal to noise ratio of 150 as in Jones et al. (2001).

Daily pressure and temperature vertical profiles were taken from the National Center for Environment Prediction (NCEP) for the actual dates and location of the measurements.

2.2 Information content analysis

The OEM allows to characterise the retrieved profiles by the use of the so-called averaging kernels, as described by Rodgers (1990, 2000). The retrieved profile x_r is related to the true profile x and to the a priori profile x_a by

$$x_r = x_a + \mathbf{A}(x - x_a) + (\text{errors}). \quad (1)$$

in which \mathbf{A} is the matrix whose rows are the averaging kernels. Eq. (1) shows that for each layer, the retrieved vmr is a weighted mean of the whole profile with weights given by the corresponding averaging kernel. In the ideal case, \mathbf{A} is the unit matrix \mathbf{I} , while in a real case the averaging kernels

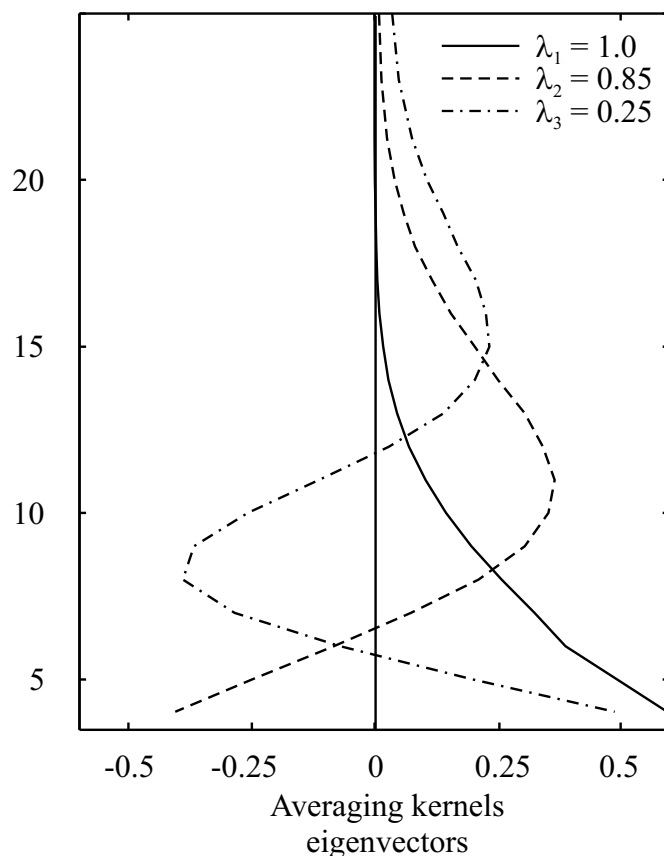


Fig. 2. Leading eigenvectors of the CO vmr averaging kernels (SZA= 61° , OPD=175 cm). The corresponding eigenvalues (λ) are given in the legend.

are bell-shaped functions. The amplitudes of these functions give an indication of the sensitivity of the retrieval in each layer, and their widths of the vertical resolution for that layer.

The averaging kernels of the retrievals are displayed in Fig. 1 for 3 km merged layers between 3.58 and 15.5 km. They show in particular that the retrievals are very sensitive to the lowermost part of the CO profile, with a vertical resolution of 3 km at the surface, degrading to 8 km above.

The decomposition of the averaging kernels matrix in eigenvectors and eigenvalues gives a more quantitative estimation of the information retrieved from the measurements. The eigenvectors corresponding to eigenvalues close to 1 are the components of the CO profile that are retrieved from the measurement, while the eigenvectors corresponding to eigenvalues close to zero are the components of the CO profile that come from the a priori information. The eigenvalues give the ratio of information coming from the measurement for the corresponding components. The number of independent pieces of information retrieved from the measurements is the trace of the averaging kernel matrix (Rodgers, 1990, 2000).

Table 1. Random error budget for the retrieval of CO vmr profiles and total columns (%) from the FTIR at ISSJ

Error source	Temperature uncertainty	Measurement error	Smoothing error	Total random error	Variability
3.58–6.5 km	5.4	2.6	2.9	6.7	25
6.5–9.5 km	2.3	3.4	5.8	7.1	20
9.5–12.5 km	1.8	3.6	4.8	6.3	19
12.5–15.5 km	0.1	3.4	6.6	7.4	22
15.5–18.5 km	0.5	3.0	10.1	10.5	20
Total Column	1.3	0.6	0.4	1.5	19

On average, our retrievals contain 2.1 independent pieces of information. The three leading eigenvectors of the averaging kernels matrix and the corresponding eigenvalues are displayed in Fig. 2. The first component is maximum at the ground and reaches its half maximum at 8.5 km, while the second component has two extrema, one at the ground and a broader one between 8 and 15.5 km. For these two components, most of the information (100% for the first component and 85% for the second one) is retrieved from the measurement. The information about the CO profiles contained in the measurements is therefore mostly comprised in the altitude range between the ground and 15.5 km. The two leading components do not represent independent layers. Nevertheless, they allow to retrieve information about the variations of CO below ~ 8 km and between ~ 8 and ~ 15.5 km almost independently. Only 25% of the information corresponding to the third component comes from the measurement. This last meaningful component allows to improve the height resolution.

2.3 Error analysis

Uncertainties associated with the CO mean vmr in the 3 km merged layers from 3.58 to 18.5 km and with the total column of CO are listed in Table 1 for the random errors and in Table 2 for the systematic errors. The measurement and smoothing errors are estimated following the formalism described in Rodgers (1990). As in Barret et al. (2002), the a priori and the measurement covariance matrices used to compute the smoothing and the measurement errors are different from the ad hoc matrices used for the retrievals. They are chosen to represent more realistically the uncertainties on the a priori CO profile and on the measurements. The measurement errors have been computed assuming an effective signal to noise ratio of 200 for the spectra. This value corresponds to the mean quality of the fit between the simulated and the measured spectra. The smoothing errors have been computed with the a priori covariance matrix used for the MOPITT operational retrievals (Deeter et al., 2003), whose variabilities have been reduced by 10% in order to account for the lower

variability of CO above the site of the Jungfraujoch. The latter was estimated from the routine in situ surface measurements performed at the Jungfraujoch (Sect. 3) and from the time series of FTIR CO total columns (Sect. 4). The resulting relative variabilities are listed in the last column of Table 1. The smoothing error takes into account the a priori profile contribution and the smoothing of the true profile by the averaging kernels. In agreement with the information content analysis, this error is therefore the smallest in the first layer (3.58–6.5 km) where the 2 leading independent retrieved components have high values, and it is the highest in the 15.5–18.5 km layer where these components have low values (Fig. 2, Sect. 2.2).

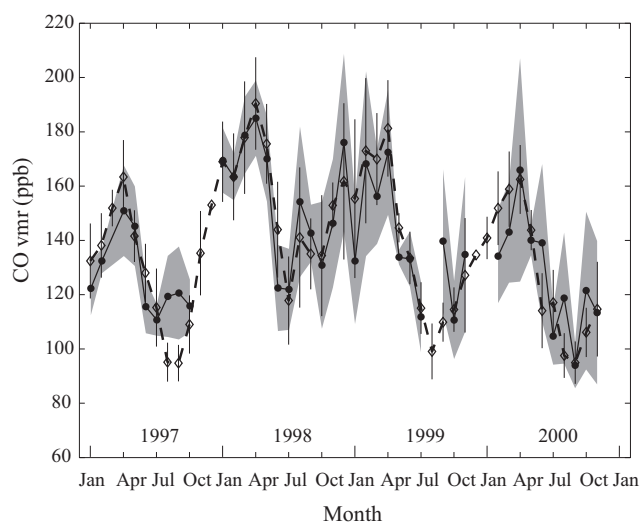
The other sources of error come from uncertainties on parameters or ancillary data used in the retrieval. They are estimated by a perturbation method (Barret et al., 2002). A spectrum synthesized using the actual ancillary data or retrieval parameter is retrieved with these same data or parameter to which their uncertainties are added. The error is estimated as the difference between the retrieved and the true profile. Following Rinsland et al. (1999) the temperature uncertainty is set to 2 K at all altitudes. For the CO absorption lines, the air broadening coefficient uncertainty is set to 2% and the line intensity uncertainty to 5% (Rothman et al., 2003). The effect of systematic distortions of the instrumental line shape (ILS) is estimated following Rinsland et al. (1999). The ILS is modeled by a straight-line apodization function which value at maximum path difference is called the effective apodization parameter (EAP) (Park, 1983). The distortions of the ILS are represented by a 10% perturbation of the EAP. The systematic error due to the limitations and approximations of the forward model is taken from Rinsland et al. (2000).

The comparison between the a priori variabilities and the total random errors shows that the uncertainties on the CO vmr's are 2 to 4 times lower after the measurement.

The total systematic errors are all comprised between 6 and 9%.

Table 2. Systematic error budget for the retrieval of CO vmr profiles and total columns (%) from the FTIR at ISSJ

Error source	Air broadening coefficient	Line intensity	Forward model approximations	ILS distortions	Total systematic error
3.58–6.5 km	6.2	5.0	4.0	1.2	9.0
6.5–9.5 km	2.8	5.0	4.0	1.3	7.1
9.5–12.5 km	5.0	5.0	4.0	1.9	8.3
12.5–15.5 km	3.3	5.0	4.0	1.2	7.3
15.5–18.5 km	1.4	5.0	4.0	0.5	6.6
Total Column	0.5	5.0	4.0	0.3	6.4

**Fig. 3.** Comparison between FTIR and in situ monthly averaged vmr's. Full line with full circles and shaded area: in situ surface vmr's and associated monthly variability (1σ). Dashed line with empty diamonds and associated error bars: FTIR 3.58–6.5 km averaged vmr and associated monthly variabilities.

3 Comparison of CO surface in situ and FTIR measurements

The Swiss Federal Laboratories for Materials Testing and Research (EMPA) performs continuous in situ surface measurements of CO vmr's at the Jungfraujoch since 1996. The overall uncertainty is estimated to be 10% on hourly mean values (Forrer et al., 2000). These surface measurements provide an opportunity to validate the retrieved FTIR vmr near the ground. We have compared the monthly averaged surface vmr with the 3.58–6.5 km layer vmr retrieved from the FTIR measurements. Only measurements for days with correlative FTIR observations are included in the monthly surface vmr averages. In order to eliminate high surface variability caused by boundary layer processes, days with surface CO vmr variability greater than 20 ppbv have been rejected in

the average. The results of the comparison are displayed in Fig. 3.

Both datasets show a very good agreement and are highly correlated (correlation coefficient of 0.87 for 41 months of comparison). The mean difference between the FTIR surface vmr's and the in situ data is (0 ± 13) ppbv. The highest discrepancies are found in the late summer months and are believed to result from thermally induced vertical transport that brings polluted air from the boundary layer to the station (Lugauer et al., 1998). The abnormally high CO concentrations detected by both measurement techniques in the summer of 1998 correspond to a 10-year CO summertime maximum observed in the Extra-Tropical Northern-Hemisphere and attributed to above average forest fires in America and in Russia (Wotawa et al., 2001).

4 Comparison of MOPITT and ground-based CO measurements

MOPITT is a nadir-sounding instrument onboard the Terra platform that has been launched on 18 December 1999. Its purpose is to measure CO and CH₄ total columns as well as CO vertical distributions with a horizontal resolution of 22 km. The instrument is described in Drummond et al. (1996), and the retrieval algorithm in Pan et al. (1998). The operational CO retrieval algorithm which produces the version 3 level 2 data used in this study is described in Deeter et al. (2003). The retrievals are based on 3 of the planned 6 instrument signal channels that exhibit a very low relative sensitivity to the boundary layer. For measurements over land, the higher thermal contrast between the surface and the atmosphere during the day makes daytime measurements more sensitive to the boundary layer than nighttime measurements. Rodgers and Connor (2003) describe how to compare MOPITT level 2 data to ground-based FTIR data by taking the different vertical sensitivities of the instruments into account. From simulated comparisons between profiles and total columns from both instruments, they conclude that it is more meaningful to compare total columns. Because of

the high altitude of the ISSJ station, total columns cannot be compared. However, using the formalism of Rodgers and Connor (2003), we are able to take the altitude of the station into account in a more quantitative way than Pougatchev et al. (1998) in their comparison of CO columns from the Measurement of Air Pollution from Space (MAPS) on board the Space Shuttle with ground-based FTIR data.

We compare the daily average FTIR column with the MOPITT column above the station, calculated as the average over all pixels satisfying the following criteria of coincidence. The pixels must lie within a circle of 300 km around the ISSJ, they must be cloud free, and their retrieval bottom pressure must be higher than 865 hPa. This last criterion used to eliminate pixels with less than 7 retrieval levels, causes a loss of less than 5% of the data. Daytime MOPITT measurements are compared with FTIR observations taken on the same day. Nighttime MOPITT measurements are compared with FTIR observations taken the day before or the day after. The assumption that the FTIR and the MOPITT CO columns probably exhibit little diurnal variations is supported by the following facts: (i) CO has a chemical lifetime of ~ 2 months (Daniel and Solomon, 1998), (ii) no diurnal variations are detected from the FTIR columns time series meaning that they are not sensitive to diurnal variations induced by convection in spring and summertime in mountainous areas (Lugauer et al., 1998; Forrer et al., 2000).

In order to take into account the difference in height sensitivity between both instruments, we must compute the column averaging kernels (Rodgers and Connor, 2003). The retrieved column c_r is computed from the retrieved profile x_r , the a priori profile x_a and the a priori column c_a using the column operator \mathbf{C} , following: $c_r = c_a + \mathbf{C}^t(x_r - x_a)$ with t indicating the transpose of the matrix. The column operator is a column vector formed of the partial air columns in each retrieval layer in molecules/cm². When \mathbf{C} is determined, the column averaging kernel, a , can be computed from the profile averaging kernel matrix \mathbf{A} as:

$$\mathbf{a}' = \mathbf{C}' \mathbf{A}. \quad (2)$$

The MOPITT CO vmr's have been interpolated on 7 pressure levels such that the pressure at the altitude of the ISSJ corresponds to the upper boundary of the third layer. The column averaging kernels appropriate for the column above the altitude of the station, normalised by the total column operator \mathbf{C} , are displayed for both instruments in Fig. 4. For the FTIR instrument, this averaging kernel is the total column averaging kernel, while for MOPITT it is a partial column averaging kernel, obtained by setting the first 3 elements of \mathbf{C} in Eq. (2) equal to zero. The FTIR total column averaging kernel is very close to the total column operator and introduces almost no smoothing. The MOPITT partial column averaging kernel introduces two kinds of smoothing error. The kernels show less sensitivity to the first and fourth layers above the ISSJ altitude than to the two middle layers. The non-zero terms in the kernels for the 3 layers below the ISSJ

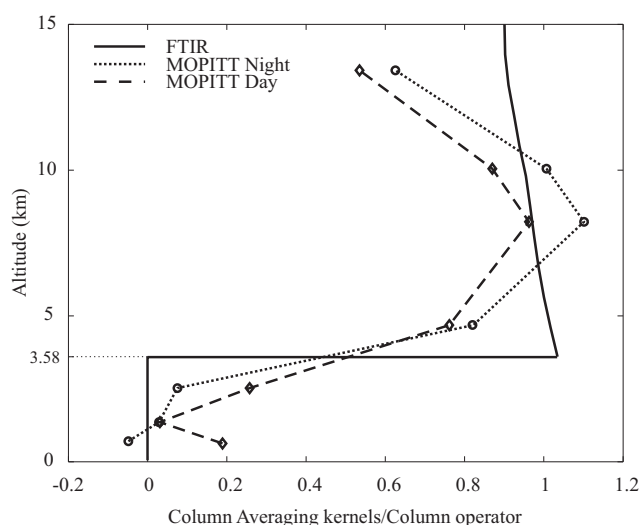


Fig. 4. Ratio of the CO total column (resp. partial column above 3.6 km) averaging kernels for the FTIR (resp. MOPITT) instrument to the total column operator. Full line: FTIR total column; dashed: MOPITT daytime; dotted: MOPITT nighttime.

Table 3. Retrieval errors for CO columns from MOPITT (in 10^{16} molecules/cm²)

	Day	Night
Total Column	7.4	15
Partial Column below ISSJ	21	35
Partial Column above ISSJ	3.4	3.5

altitude are responsible for bringing some information from the profile below this altitude into the partial column above it.

The retrieval errors computed according to Rodgers (2000) are summarized in Table 3. It shows that the MOPITT error is reduced significantly when considering partial columns above the ISSJ instead of total columns.

In order to quantify the impact of the column averaging kernels smoothing effects, we have simulated the MOPITT columns above the ISSJ using the FTIR retrieved profiles, hereinafter indicated as c_{FM} , as in Eq. (25) in Rodgers and Connor (2003). Above the ISSJ altitude, the retrieved FTIR profiles have been interpolated on the 4 upper modified MOPITT pressure levels. On the 3 lower levels, we used the CO vmr's retrieved from MOPITT as a proxy for the missing FTIR values. This is shown explicitly in Eq. (3), in which the subscript F stands for FTIR and M for MOPITT, and the level numbering refers to the modified MOPITT pressure levels.

$$c_{FM} = c_a + \mathbf{a}'_{M(4:7)}(x_{rF(4:7)} - x_{a(4:7)}) + \mathbf{a}'_{M(1:3)}(x_{rM(1:3)} - x_{a(1:3)}) \quad (3)$$

Table 4. Statistics of the comparison between FTIR and MOPITT CO columns (in 10^{16} molecules/cm²). c_F stands for FTIR total column, c_M for MOPITT partial column above ISSJ and c_{FM} for FTIR total columns smoothed by the MOPITT partial column averaging kernel; $R(x, y)$ is the correlation coefficient between quantities x and y ; Δc_{FM} as defined in Eq. (4) is taken into account in the standard deviations of $c_{FM} - c_M$

	Daytime	Nighttime
Number of days	41	42
$c_F - c_M$	-8.4 ± 5.8	-1.0 ± 7.5
$R(c_F, c_M)$	0.92	0.87
$c_{FM} - c_M$	-8.1 ± 7.7	-3.8 ± 7.4
$R(c_{FM}, c_M)$	0.89	0.87
Δc_{FM}	± 3.4	± 0.4

The impact of using the MOPITT values on the 3 lower levels is quantified as:

$$\Delta c_{FM} = \mathbf{a}_{M(1:3)}^t \sigma_{rM(1:3)} \quad (4)$$

where σ_{rM} is the MOPITT retrieval error profile (see Table 4). Comparisons between MOPITT and FTIR data are more straightforward when the instrument is located at sea level because no approximation for the FTIR values is needed in this case. Figure 5 displays the comparisons between FTIR and MOPITT columns; the corresponding statistics are presented in Table 4. For both daytime and nighttime measurements, MOPITT overestimates the CO columns relative to the FTIR. The nighttime/daytime MOPITT column differences relative to FTIR columns are reduced when the smoothing of Eq. (3) is applied to the FTIR profiles. The difference of sensitivity between night- and daytime MOPITT measurements may therefore be partly responsible for the night/day discrepancy. A more significant effect is expected for FTIR instruments located at sea level because the difference in night/day sensitivity is highest in the lowermost levels (Deeter et al., 2003).

The amplitude of the CO seasonal variations, quantified by fitting a single sinusoid with a period of one year through the time series of FTIR CO total columns from January 1999 to May 2001, is 16.5%. The standard deviations of the FTIR MOPITT differences are therefore at least 2 times lower than the seasonal variations, showing that both instruments reproduce the CO variations correctly. This is confirmed by the correlation coefficients that are all close to 0.9 (Table 4).

5 Conclusions

The quality of the CO vertical profile information extracted from ground-based FTIR solar absorption observations at the Jungfraujoch has been demonstrated. The measurements contain ~ 2 independent pieces of information that allow

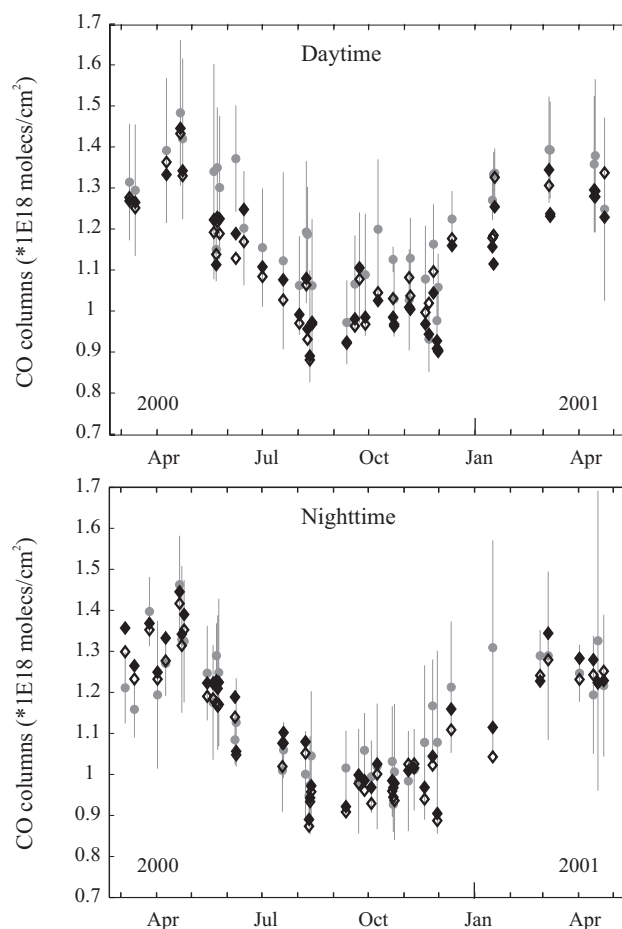


Fig. 5. FTIR and MOPITT daily averaged columns. Upper plot for daytime, lower plot for nighttime MOPITT measurements. Full diamonds: FTIR total columns; empty diamonds: FTIR total columns smoothed with MOPITT kernels (Eq. 2); full gray circles: MOPITT partial columns above the ISSJ altitude; gray error bars represent the MOPITT variability (1σ) for all the pixels entering the daily average (see text for details).

to retrieve the CO abundances in the lower to middle troposphere (3.58–8 km) and in the upper-troposphere-lower-stratosphere (8–15 km) almost independently. In 3 km thick layers from the ground (3.58 km) to 15.5 km, the CO vmr is retrieved with precisions and accuracies better than 10%. The monthly means of the average vmr in the 3 km layer above the altitude of the station follow closely the monthly means of the correlative in situ surface measurements at the station without significant bias.

Comparisons between the CO columns above the ISSJ derived from the FTIR and from the MOPITT space borne instrument, taking into account the smoothing affecting MOPITT columns, show that MOPITT values are $8 \pm 8\%$ (resp. $4 \pm 7\%$) higher than the corresponding FTIR values for daytime (resp. nighttime) MOPITT measurements. While CO seasonal variations are estimated from the FTIR data to be of

the order of $\pm 16\%$, this comparison proves that MOPITT is able to provide useful geophysical information about the free tropospheric CO at northern mid-latitudes.

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